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Variables of Dynamics in Supercooled liquids

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凝縮系物理学における重要な未解決問題である過冷却液体のガラス転移の解明には過冷却液体のダイナミクスの理解が欠かせない。ガラス転移に向かう冷却の過程で過冷却液体の粘性やα緩和時間などが急激に増大するが、それにはダイナミクスの多様化を伴う。典型的な例が誘電分光で観測されるα過程とJohari-Goldstein(JG)-β過程の分岐で、これは過冷却液体に普遍的な現象である。我々は、分岐現象を含む糖アルコール系のダイナミクスを広帯域誘電分光により詳細に調べた。その結果、過冷却糖アルコール系におけるこれらのダイナミクスは、水分率や平均分子量など温度以外の変数で再現できることが分かった。

Liquid-glass transition is one of the important unsolved problems in condensed matter physics. Liquid undergoes the glass transition through the supercooled liquid state. On the other hand, supercooled liquid follows a sub-stable state off the free-energy minimum in contrast to non-equilibrium glassy state. Therefore, understanding supercooled liquid must be essential to reveal the nature of the glass transition.

The most remarkable feature of the supercooled liquid can be found in its dynamics. Dynamical features of the supercooled liquid change drastically in cooling toward the glass transition temperature. As temperature decreases, the viscosity and the dielectric α relaxation time increase in non-Arrhenius manner. This variation is accompanied by diversification of microscopic dynamics. Typical example of the diversification is the bifurcation of the α and the JG-β processes observed in dielectric relaxation spectroscopy. Since this bifurcation is a common feature of supercooled liquid, some theoretical approaches have been examined to explain that [1].

We have studied dynamics including this bifurcation in sugar alcohol systems by means of broadband dielectric spectroscopy. It has been reported that adding small amount of water into sorbitol makes the α relaxation time faster while the β relaxation time not so much [2]. This effect is similar to the size effect of component molecules [3]. These facts suggest that the dynamics

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in supercooled liquid can be controlled not only by temperature but also other parameters such as water content in sugar alcohols.

Figure 1 (a) shows the plot of relaxation frequency with maximum dielectric loss against reciprocal temperature indicating the typical bifurcation behavior for anhydrous sorbitol. On the other hand, similar plot is given in Fig.1(b). In this case, however, the relaxation frequencies of water-in-sorbitol mixtures are plotted against sorbitol molar fraction, x_{sor}, at a constant temperature (264K). A surprising finding is the similarity of loci in these figures. This finding suggests that "supercooling" of a sugar alcohol with water system can be brought not only by thermal quenching but also removing of water. It seems that this idea can be applied to many other systems especially those in life.

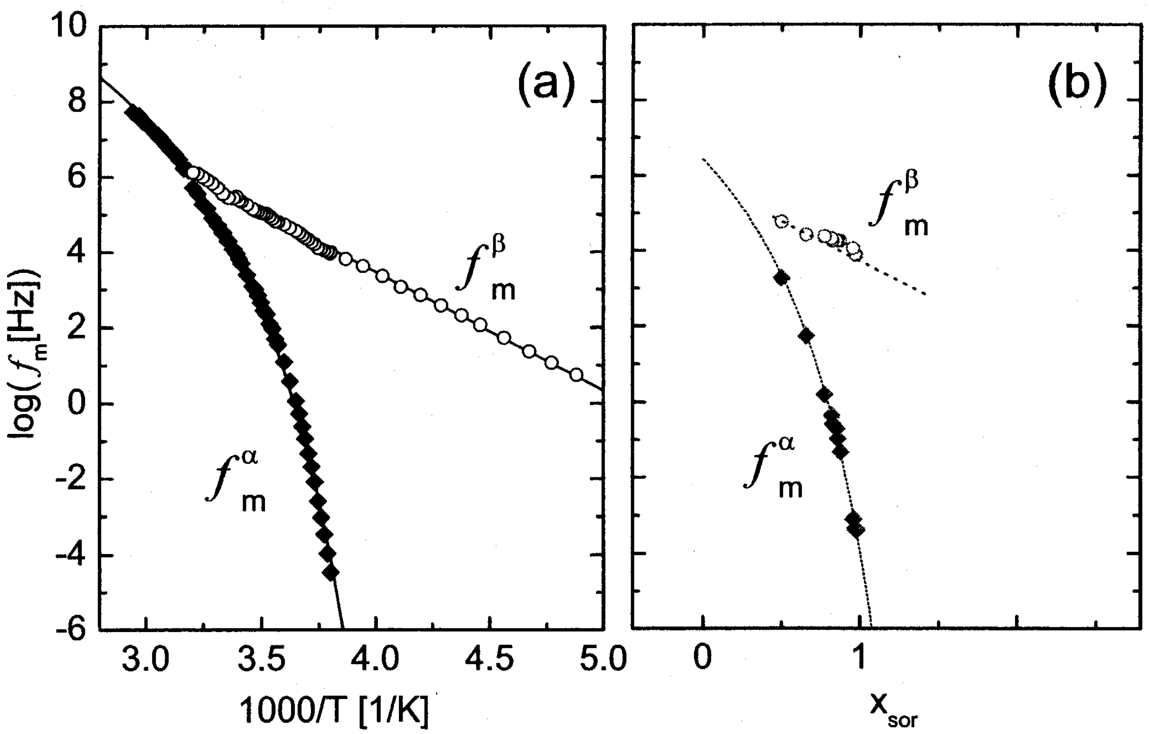

Fig.1 Plots of logf_m of the α and the JG-β processes versus (a) 1/T and (b) x_{sor}.

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